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Study of Hydrodynamics Factor (Off-Bottom Clearance) in Enhancing Hydrogen Gas Production as a Renewable Energy in a Stirred Bioreactor

Wa Ode Cakra Nirwana[a], Septila Renata[a], Esterina Dian Anggraini[a], Tantular Nurtono[a*], Sugeng Winardi[a]

[a]Department of Chemical Engineering
Institute of Technology Sepuluh Nopember, Surabaya 60111 Indonesia

Abstract

Hydrogen has been recognized as an ideal energy source that is environmentally friendly and widely used in various industries. Burning hydrogen does not pollute the environment, can be produced from renewable materials and produce a higher energy yield compared to hydrocarbon fuels. During this time, hydrogen production process by fermentation (biohydrogen) more review in terms of chemical and biological factors, whereas physical factors such as hydrodynamic factors are rarely studied. Hydrodynamic factor is one of the important factor affecting the bio-hydrogen production processes involving multiphase system. Therefore, this research aims to study the influence of hydrodynamic factor that is off-bottom clearance (position of the impeller from the bottom of the tank) influence to the hydrogen production process by fermentation. Off-bottom clearance is one of the important hydrodynamic factor because it can affect the flow pattern and then can affect the distribution of microbes in the media and mass transfer of hydrogen gas from liquid phase to the gas phase. Hydrogen was produced using glucose as substrate and Enterobacter aerogenes (NBRC 13534) as the bacteria. The reactor used is a stirred tank equipped with a single impeller, 45^o-6 blades, Pitched Blade Turbine (PBT) type at constant rotation speed of 200 rpm. While off bottom clearance was varied 0.25 H, 0.5 H and 0.75 H. The results showed that at the same impeller speed, impeller position does not affect to the speed of consumption of glucose by bacteria that are used as a carbon source for metabolism, but the position of the impeller effect on the capacity of hydrogen gas. A lower off bottom clearance provides better performance in an effort to increase the hydrogen production capacity. The highest yield was 0.0029 mol H_2/mol glucose, which was obtained at impeller position 0.25 H.

Keyword: *hydrodynamics, biohydrogen, off bottom clearance, pitched blade turbine*

1. Introduction

Fossil fuels as a primary energy source are a finite resource. Currently, the world's energy requirement is still very dependent on fossil energy, therefore the development of alternative energy becomes an urgent need. This alternative energy should be environmentally benign, low emission and renewable. Among the development of sustainable energy sources, hydrogen is a good candidate as a future energy source since it has low emission, cleaner, renewable and environmentally benign.

Hydrogen is an ideal alternative fuel since it produces only water when burning, generates higher energy yield (122kJ/g) by 2.75 times than hydrocarbon fuels (Kim et. al., 2006) and could be directly used to produce electricity through fuel cells (Li et. al., 2008). Moreover, hydrogen as a new energetic vector can be stored and transferred as a chemical energy (Aceves-Lara et al., 2008). At present, hydrogen mainly produced from the physicocemical process, include steam reforming-natural gas, electrolysis, partial oxidation of fuel oil, gasification of coal, gasification of biomass and so on. These processes require external energy sources, so these are uneconomic (Kim et al., 2004). Hydrogen production biologically is become a viable method since it offers the possibility to use low-price renewable feedstock.

Bio-hydrogen production from various types of substrates that have been widely studied by previous researchers are generally more focused on studying the biological and chemical factors that can affect the efficiency of hydrogen production. Whereas, bio-hydrogen production is a complex process involving chemical, biological and physical processes. A number of chemical and biological factors that can influence the efficiency of hydrogen production such as fermentation type, microorganisms type, the influence of different substrates, the influence of pH and substrate concentration effect has been widely studied (Yokoi et al., 1995; Chang et al., 2002, Liu et al., 2004; Nath and Das, 2004; Li et al., 2008; Ruggeri et al., 2009). Physical characteristics such as hydrodynamics factors affecting biohydrogen production efficiency is still rarely studied (Aceves-Lara et al., 2008; Chou et al., 2008; Gómez et al., 2009; Wang et al., 2009 and Ding et al ., 2010), whereas the fermentation process that occur in a stirred tank is a multiphase system that requires hydrodynamics review. Moreover, it has also attempted to study the optimal reactor configuration to increase the hydrogen yield (Lee et al., 2006; Lo et al., 2009; Distefano et al., 2010). Therefore, the purpose of this research is to study the influence of hydrodynamics factor that is off-bottom clearance

*Corresponding Author's E-mail: tantular@chem-eng.its.ac.id

(position of the impeller from the bottom of the tank) influence to the hydrogen production process by fermentation. Off-bottom clearance is one of the important hydrodynamics factor since it can affect the flow pattern and then can affect the distribution of microbes in the media and mass transfer of hydrogen gas from liquid phase to the gas phase.

In this study, hydrogen production by anaerobic fermentation performed in a stirred tank reactor with impeller type Pitched Blade Turbine (PBT), blade angle of 45^o and the number of blade 6. PBT impeller types can produce axial and radial flow patterns and provide upward and downward flow direction depending on the direction of rotation. In addition, PBT impeller also has the lowest power number compared with other turbine impeller types such as disc turbine, straight blade turbine and a curved blade turbine (Tatterson, 1991). Shear level generated by this impeller type is low with very high flow level. PBT has a small difference in the relative velocity between the blade and the surrounding fluid so that at the same impeller speed, PBT produces shear rate smaller than the flat blade impeller (Tatterson, 1994). It provides an advantage because the bacteria are very susceptible to mechanical treatment.

2. Theory

Many types of substrate can be fermented to produce hydrogen such as glucose, hexose isomers, glycerol, starch or cellulose but glucose is the most commonly used. Glucose is a carbon source that is easy to digest by microorganisms and present in most agricultural waste. Glucose can be fermented to produce hydrogen with varies amount depending on the fermentation method and final product based on the following reaction (Chou et.al., 2008):

$$C_6H_{12}O_6 + 2H_2O \xrightarrow{\text{mikroba}} 2CH_3COOH + 2CO_2 + 4H_2$$

$$C_6H_{12}O_6 \xrightarrow{\text{mikroba}} CH_3CH_2CH_2COOH + 2CO_2 + 2H_2$$

As described before, glucose fermentation process to produce hydrogen is a process that involve multiphase system. H_2 partial pressure in the liquid phase is one of the key factors affecting H_2 production. The effects of poor mass transfer and H_2 inhibition can be crucial for solid wastes converting into H_2 in bioreactors (Chou et.al., 2008). Therefore, an effort to reduce the H_2 solubility in liquid phase is needed, thus H_2 gas can release to the gas phase and leave reactor easily. Off-bottom clearance is suspected to play a role in determine the releasing of hydrogen gas from the liquid phase. Generally, off-bottom clearance can affect the overall flow pattern in a stirred tank for producing radial and axial flow. In other words, the flow pattern will change as a function of clearance. At a low clearance, the circulation patterns of the PBT impeller will turn into a radial flow (Tatterson, 1991).

Hydrodynamics study in a bioreactor can be conducted experimentally and computationally. When fluid dynamic analyzing in bioreactor is needed, experiment method become uneconomic, can not give an accurate information to understand fluid dynamic in micro scale and it can not describe the whole phenomena that occur in the whole of the tank. Therefore, computational approach is needed that can give more detail information and turbulence characteristics of the stirred tank reactor in various operation condition and tank geometry. Computational method is more economic, faster, can give a good approximation to predict the hydrodynamic in a system and can be used to visualize detailed flow phenomena when the direct measurement of parameters such as pressure, velocity and phase volume fraction can be difficult (Wang et. al., 2010). Computational Fluid Dynamic (CFD) is the most popular method to analize the characteristic of bioreactor.

3. Materials and Methods

3.1. Medium

Enterobacter aerogenes NBRC 13534, from Prof. Hiroyasu Ogino, *Osaka Prefecture University* at Japan was grown on potatoes dextrose agar (PDA) medium containing glucose, OHLY yeast extract, potatoes extract and agar. The fermentation was conducted in two stages. The first stage is the stage of acclimatization in a container was filled to 2 L of solution containing the following components (in 2 L of distilled water): 40 g of glucose, 10 g of OHLY yeast extract, $C_{12}H_{24}FeO_{14}$ 250 mg, $MnSO_4$ 0,2 mg, $CuSO_4$ 0,2 mg. The second stage is the stage of fermentation in the bioreactor with a working volume of 14.5 L (consisting of per L of distilled water: glucose 2% w/v and OHLY yeast extract 0.5% w/v).

3.2. Reactor system

The reactor was employed for batch fermentation with a working volume of 14.5 L. Inside diameter (T) of 0.21 m and height (H) 0.45 m and it was equipped with a stirring system made of 6-blade 45^o Pitched Blade Turbine (PBT). The dimensions of the impeller is diameter (D) of 0.072 m, width of blade (0.2 D) = 0.015 m. In addition, the reactor wall is also fitted with four baffles of width 0.022 m and thickness 0.003 m. In order to determine the influence of hydrodynamic on the hydrogen production, the clearance (C) was varied that is 0.25 H, 0.5 H, and 0.75 H and the stirring velocity constant at 200 rpm with the direction of rotation is pumping down. The pH was kept on 5.5 – 6.5 by adding NaOH 4 M and the temperature was maintained constant at 37 ^{o}C. Schematic of the reactor and dimension of the impeller used in the present study can be seen in Figure 1.

3.3. Analitycal method

The glucose concentration was analyzed using a spectrophotometer (Genesys 10uv, Thermo Scientific) at a wavelength of 510 nm. Before analyzing, the

sample was added glucose reagent (Diagnostics Media SRL) and incubated at 37^{o}C for 5 minutes. Bacterial cell was determined using a spectrophotometer (Genesys 10uv, Thermo Scientific) at a wavelength of 600 nm and calibrated by manual calculation using counting chamber. The pH values were measured by a pH-meter (pHmeter AZ8601). The gas flow rate was measured with a flowmeter and the hydrogen concentration was measured on-line by hydrogen analyzer (H_2 Panterra Sensor, Neodym).

3.4. Computational Approach for Bioreactor System

The geometry of bioreactor has analyzed with a computational three dimensional mesh. The meshes were created in the ANSYS Fluent Meshing and exported into the ANSYS Fluent 12.1. Computational approach used to predict and visualize the flow pattern of fluid (assumed as water) and the gas tracking that assumed as hydrogen gas only in the bioreactor. Hydrogen was considered as spherical particles with a constant diameter of 1 mm. The hydrogen particles were injected in the bioreactor at ten points with maximum number of step of 500000.

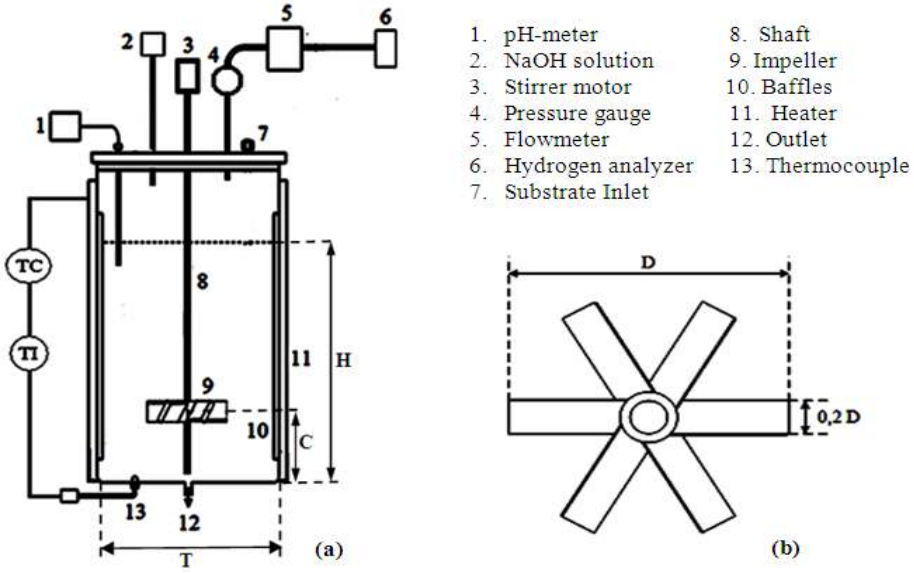

Figure 1. Schematic view of the reactor (a) and dimension of impeller (b)

4. Result and Discussion

4.1. Effect of Clearance on Degradation of Glucose Concentration

Glucose analysis was conducted at the early fermentation process (when the substrate was inserted into the reactor) and at the end of the fermentation (when the hydrogen gas was not detectable by hydrogen analyzer) using a spectrophotometer at a wavelength of 510 nm. Figure 2 shows that the position of the impeller do not give effect on the degradation of glucose concentration. When glucose concentration measured at the end of the fermentation, it showed that glucose concentration reached almost 0 g/L at the different impeller clearance. It means that at the same impeller speed, impeller clearance do not affect on the consumption of glucose by bacteria that are used as a carbon source for metabolism. Glucose will be consumed completely even in the different time period for each impeller position and the bacteria will be stopped producing hydrogen only if glucose content in the medium has run out.

Profile of degradation of glucose concentration each time also can be seen in Figure 2. The initial glucose concentration differ for each clearance because the initial glucose concentration measured when all the substrate was inserted into the reactor so the condition of each experiment may vary. In 4 hours early, the glucose degradation were extremely close to 0 g/L (at clearance of 0.75 H). Then the glucose concentration constant and finally completely consumed after the fermentation process occured for 14 hours. At clearance of 0.25 H and 0.5 H, degradation of glucose concentration occurs gradually until finally completely consumed at the 23rd hour for clearance of 0.25 H and 26th hour for clearance of 0.5 H.

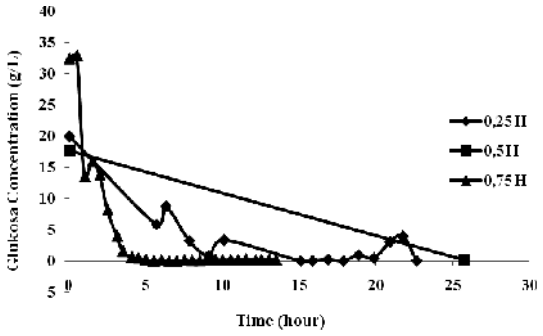

Figure 2. Degradation of glucose concentration during fermentation process at various impeller clearance

4.2. Effect of Clearance on Gas Volume Produced

As explained previous that the position of the impeller from the bottom of the tank affect the flow pattern in the stirred reactor (Figure 3). If the distance between the impeller to the bottom of the tank greater, the turbulence intensity decrease (Tatterson, 1991). It is due to for pumping down flow, at a low clearance, flow generated by impeller rotation will hit the bottom of the tank, therefore fluid have more power to flow upward axially. While at a high clearance, the fluid that pulled down less able to resist the flow on it so that the turbulence intensity decreased. Differences on flow patterns also influence the volume of gas that formed, as shown in Figure 4. The reduction in total gas volume which is formed also followed by the reduction in the volume of hydrogen produced. It can be seen that at 0.25 H clearance, the volume of gas produced of 4229.6 ml and then decrease to 462 ml at 0.5 H clearance, and at 0.75 H clearance increase to 660 ml. Enhancement of gas volume at the clearance of 0.75 H can be caused by the impeller position was closer to the liquid surface so the influence of the flow of the upper impeller is smaller than at 0.5 H clearance as can be seen in Figure 3. Therefore, the gas that formed will be discharged from the liquid phase easier than at clearance of 0.5 H.

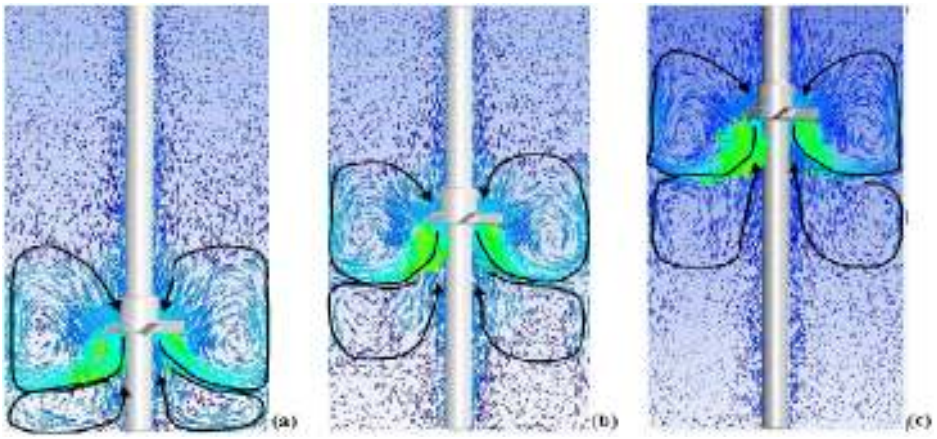

Figure 3. Fluid flow pattern at 0.25 H (a), 0.5 H (b), 0.75 H (c)

The experiment result can be supported by the simulation of H_2 particle tracking (Figure 5) where it shows the same result that at clearance of 0.25 H, residence time H_2 particles in reactor is shorter than the other clearances that is about 16.8–64.6 seconds (data not shown). It means H_2 gas is more easy to escape from liquid phase and then leave the reactor, therefore the H_2 gas volume increased. The residence time of H_2 gas at clearance of 0.5 H and 0.75 H are 12.6–155.8 seconds and 12.6–170.5 seconds, respectively (data not shown). Based on that data, residence time of H_2 at 0.5 H is shorter than at 0.75 H but at clearance of 0.5 H, not all H_2 particles are injected can be escaped from the reactor. Based on the simulation result, there are eight particles only that can be escaped (data not shown). The differrent phenomena are shown at the clearance of 0.75 H. All particles are injected can be escaped although the residence time H_2 in reactor is longer than at clearance of 0.5 H. Therefore the gas volume that released is more than gas volume at 0.5 H.

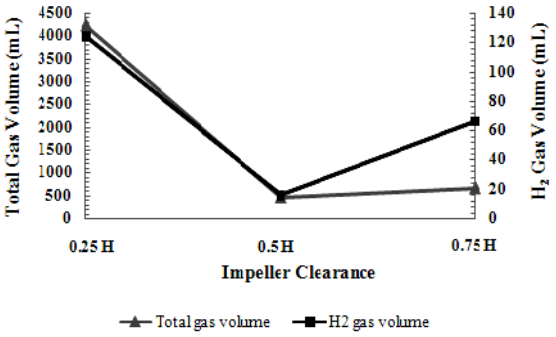

Figure 4. Effect of clearance on the total gas volume and H_2 gas produced

Figure 6 shows the accumulated gas volume generated fermentation process, where the fermentation is considered finished if the hydrogen gas was not detectable. It can be seen that the bacteria did not take long time to start producing gas. At clearance of 0.25 H and 0.5 H, bacteria need ± 1 hour only for adaptation to their enviroment (lag phase). It is indicated by the absence of gas produced by bacteria at the first hour early. While the clearance of 0.75 H, lag phase is very short because at early fermentation process, bacteria can be produced gas directly as one of the product of metabolism process.At the first hour early, the highest gas volume achieved at clearance of 0.75 H but then enhancement of gas volume formed is not significant. While at clearance of 0.25 H, gas volume increases significantly. Based on Figure 6, can be seen also that the fermentation process occured ± 24 hours, except at clearance of 0.75 H. It was correspond to the degradation of glucose concentration in the medium in the 4 hours early of fermentation process (Figure 2), which cause the bacteria did not have carbon source that can be used to produce the metabolism products, including the hydrogen gas.

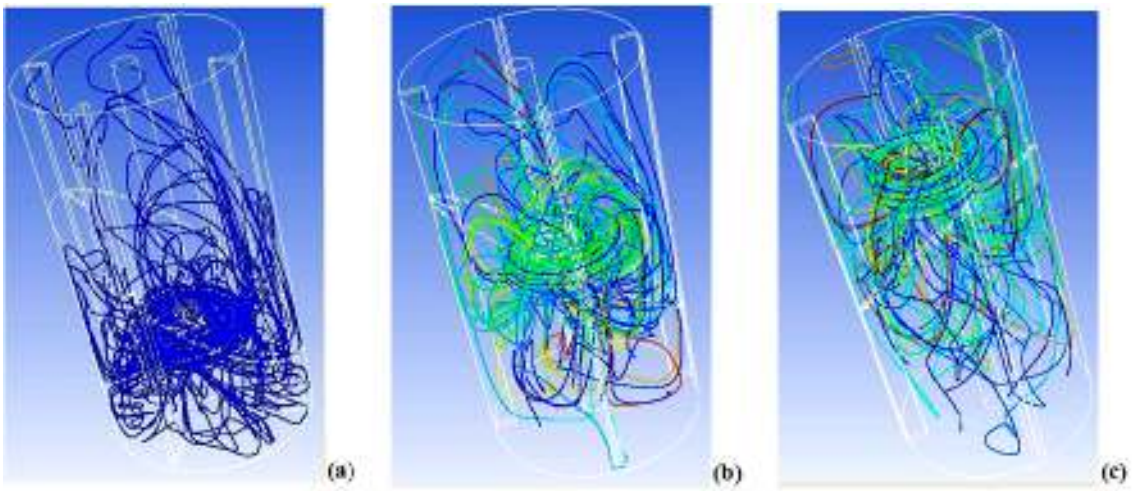

Figure 5. Hydrogen particles tracking at 0.25 H (a), 0.5 H (b), 0.75 H (c)

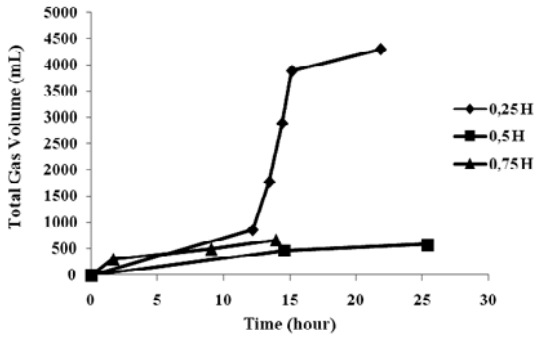

Figure 6. Total gas volume during fermentation process at various impeller clearance

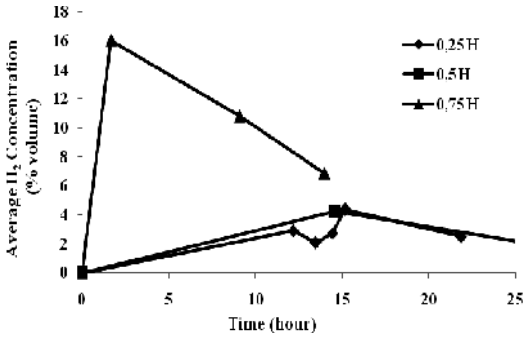

Figure 7. Average H_2 concentration during fermentation process at various impeller clearance

4.1. Effect of Clearance on Hydrogen Gas Yield

Figure 7 shows the average H_2 concentration at various impeller position. At clearance of 0.75 H, the average H_2 concentration generated at the early of fermentation is high enough. It is related to glucose consumption rate by bacteria, as shown in Figure 2, where at that impeller position, bacteria consume glucose very quickly thus bacteria need a shorter time to produce H_2.

H_2 yield obtained do not influenced by number of bacteria. Based on Figure 8, it can be seen that at the clearance of 0.25 H, number of cell bacteria is smaller than other but generate H_2 volume most widely. The yield of hydrogen produced depends only on the concentration of hydrogen and also the volume of gas produced. It is because all the glucose dissolved in the media is converted entirely. Figure 9 shows the relationship between the impeller clearance on H_2 yield. Although the highest concentration of H_2

achieved at clearance of 0.75 H and the lowest at 0.25 H but the highest hydrogen yield obtained at the clearance of 0.25 H that is 0.0029 mole H_2/mole glucose converted and the lowest is 0.00042 mole H_2/mole glucose converted at the clearance of 0.5 H. It is related to the total gas volume produced. As shown in Figure 6, the highest total gas volume was achieved at 0.25 H clearance. The H_2 yield obtained is still much lower compared to ideal yield that is 4 mole H_2/ mole glucose converted. It influenced by many factors, such as the influence of nutrition, treatment during the fermentation process, the characteristics of microorganisms that are always changing, the possibility of contamination by other microorganisms and other external factors.

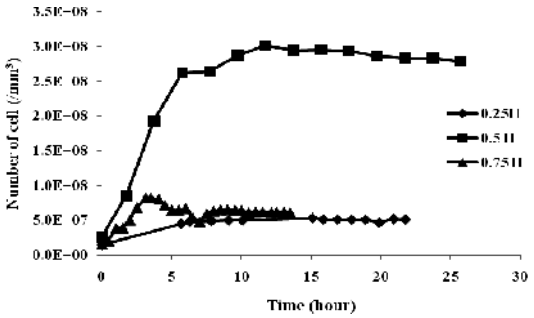

Figure 8. Effect of clearance on the number of cell

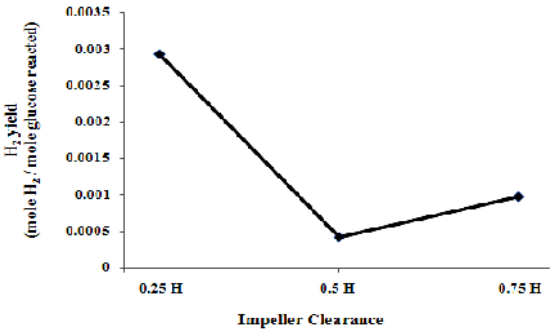

Figure 9. Effect of impeller clearance on yield H_2 produced

5. Conclusion

At the same impeller speed, off-bottom clearance do not affect on the speed of the degradation of glucose and number of bacteria but it gives significant effect on the residence time of gas in reactor that can be influenced the capacity of hydrogen gas. A lower off-bottom clearance provides better performance in an effort to increase the hydrogen production capacity since it can influence the turbulence intensity. The highest yield was 0.0029 mole H_2/mole glucose, which was obtained at clearance of 0.25 H.

6. Acknowledgment

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